Simultaneous Multiphoton-Multiatom Processes in Atomic Gases and Their Application in Enhancing Ultraweak Atomic Absorption Transitions

Yongle Yu*

State Key Laboratory of Magnetic Resonance and
Atomic and Molecular Physics, Wuhan Institute of Physics
and Mathematics, Chinese Academy of Science,
West No. 30 Xiao Hong Shan, Wuchang, Wuhan, 430071, China

Abstract

We investigate simultaneous multiphoton-multiatom (MPMA) processes in atomic gases subjected to laser fields. Our study reveals that the composite factor governing the transition rate of these processes can reach extraordinarily high magnitudes, with an intrinsic regulation mechanism causing the rate to exhibit near-saturation behavior. By integrating an MPMA process into an ultraweak atomic absorption transition, a substantial enhancement of the overall transition rate can be achieved. This enhancement enables the detection of transitions that would otherwise remain undetectable, thereby opening new avenues for exploring ultraweak quantum phenomena in atomic systems.

 $^{^*}$ yongle.yu@wipm.ac.cn

Detecting ultraweak phenomena with extremely low probabilities is a fundamental challenge in contemporary physics, essential for validating core theoretical postulates. Noteworthy among these endeavors are the detection of neutrinos [1] and the observation of atomic two-photon absorption events [2]. Neutrinos, distinguished by their exceptionally weak interactions with matter, require expansive detection volumes for observation. Similarly, atomic two-photon absorption—where an atom simultaneously absorbs two photons—occurs with negligible probability under natural conditions, but becomes observable through laser-induced enhancements. In this paper, we introduce a novel quantum amplification mechanism that has remained elusive until recently. We examine a simultaneous multiphoton-multiatom (MPMA) process in an atomic gas [3–12], demonstrating its capability to enhance ultra-weak atomic absorption transitions. Some fundamental mechanisms and unusual properties of the MPMA process have been recently elucidated in our work [12].

Consider a non-interacting atomic gas exposed to a laser field. This gas consists of two atomic species, labeled A and B. For simplicity, both A-species and B-species atoms are modeled as two-level systems, where the upper level is accessible from the lower level through an electric dipole transition. For an A-species atom, let the lower level (ground state) energy be ε_g^a and the upper level (excited state) energy be ε_e^a . The angular transition frequency is given by $\omega_a = (\varepsilon_e^a - \varepsilon_g^a)/\hbar$, where \hbar is the Planck constant. Similarly, for a B-species atom, the transition frequency is $\omega_b = (\varepsilon_e^b - \varepsilon_g^b)/\hbar$, where ε_e^b and ε_g^b represent the upper and lower level energies, respectively.

Assume the angular laser frequency $\Omega_{\mathfrak{L}}$ is set to satisfy the relation: $m\hbar\Omega_{\mathfrak{L}}=\hbar\omega_a+(m-1)\hbar\omega_b$, where m is an integer greater than 2. In this scenario, a single atom cannot be excited by absorbing a laser photon alone when $\omega_a \neq \omega_b$. However, in a high-order Quantum Electrodynamics (QED) process, an m-atom system comprising an A-species atom and (m-1) B-species atoms, can be collectively excited when each atom absorbs a laser photon simultaneously. This phenomenon is known as the multiphoton-multiatom excitation (MPMA) process. A schematic plot of a three-photon-three-atom process is depicted in Fig. 1.

In this m-atom system, the (m-1) B-species atoms are distinctly labeled as b_i $(i=1,2,\ldots,m-1)$. The Hamiltonian for the combined system, which includes these m atoms

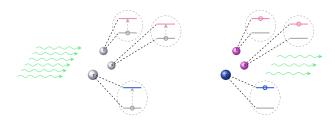


FIG. 1. Simultaneous joint excitation of a three-atom system, where each atom absorbs a laser photon.

and the laser field, is given by:

$$\hat{H} = \varepsilon_g^a |g_a\rangle\langle g_a| + \varepsilon_e^a |e_a\rangle\langle e_a| + \sum_{1 \le i \le m} (\varepsilon_g^b |g_{b_i}\rangle\langle g_{b_i}| + \varepsilon_e^b |e_{b_i}\rangle\langle e_{b_i}|) + \hbar\Omega_{\mathfrak{L}}\hat{a}\hat{a}^{\dagger} + \hat{H}_{int}, \qquad (1)$$

where $|g_a\rangle$ ($|e_a\rangle$) denotes the ground (excited) state of the A-species atom, and $|g_{b_i}\rangle$ ($|e_{b_i}\rangle$) denotes the ground (excited) state of the b_i atom. Here, \hat{a} (\hat{a}^{\dagger}) represents the annihilation (creation) operator of the laser photon, and \hat{H}_{int} describes the coupling between the atoms and the laser field:

$$\hat{H}_{int} = \hat{\mathbf{d}}_a \cdot \hat{\mathbf{E}} + \sum_i \hat{\mathbf{d}}_{b_i} \cdot \hat{\mathbf{E}}, \tag{2}$$

where $\hat{\mathbf{d}}_a$ and $\hat{\mathbf{d}}_{b_i}$ are the dipole moments of the A-species atom and the b_i atoms, respectively, and $\hat{\mathbf{E}}$ is the electric field operator of the laser.

Let the quantum state of the joint system with m atoms in the ground states and N_{γ} laser photons be denoted as $|\Psi_{i}\rangle = |g_{a}\rangle|g_{b_{1}}\rangle|g_{b_{2}}\rangle\dots|g_{b_{m-1}}\rangle|N_{\gamma}\rangle$. The quantum state with m atoms being excited by absorbing m photons cooperatively is denoted as $|\Psi_{f}\rangle = |e_{a}\rangle|e_{b_{1}}\rangle|e_{b_{2}}\rangle\dots|e_{b_{m-1}}\rangle|N_{\gamma}-m\rangle$ $(N_{\gamma}\gg m)$. The transition rate from $|\Psi_{i}\rangle$ to $|\Psi_{f}\rangle$ in the m-th order perturbation is given by:

$$W_{mpma} = \frac{2\pi}{\hbar} |T_{fi}|^2 \rho(E_f)|_{E_f = \varepsilon_e^a + (m-1)\varepsilon_e^b}.$$
 (3)

Here, $\rho(E_f)$ is the density of states of the m-atom system at energy E_f [13], and

$$T_{fi} = \sum_{\kappa_1} \sum_{\kappa_2} \cdots \sum_{\kappa_{m-1}} \frac{\langle \Psi_f | \hat{H}_{int} | \Psi_{\kappa_{m-1}} \rangle \dots \langle \Psi_{\kappa_2} \hat{H}_{int} | \Psi_{\kappa_1} \rangle \langle \Psi_{\kappa_1} | \hat{H}_{int} | \Psi_i \rangle}{(\varepsilon_{\Psi_{\kappa_{m-1}}} - \varepsilon_{\Psi_{\kappa_{m-2}}}) \dots (\varepsilon_{\Psi_{\kappa_3}} - \varepsilon_{\Psi_{\kappa_2}}) (\varepsilon_{\Psi_{\kappa_2}} - \varepsilon_{\Psi_{\kappa_1}}) (\varepsilon_{\Psi_{\kappa_1}} - \varepsilon_{\Psi_i})}, \quad (4)$$

where Ψ_{κ_j} is the virtual intermediate quantum state of the system at the j-th step in this m-th order process with an energy of $\varepsilon_{\Psi_{\kappa_j}}$, and the virtual transition path is $|\Psi_i\rangle \to |\Psi_{\kappa_1}\rangle \to |\Psi_{\kappa_2}\rangle \to \cdots \to |\Psi_{\kappa_{m-1}}\rangle \to |\Psi_f\rangle$. It is clear that Ψ_{κ_j} corresponds to a quantum state with j

atoms excited and with a laser photon number of $N_{\gamma} - j$; otherwise, its contribution to the transition vanishes. Notably, there are m! possible excitation pathways corresponding to different choices of intermediate states $|\Psi_{\kappa_1}\rangle, |\Psi_{\kappa_2}\rangle, \dots, |\Psi_{\kappa_{m-1}}\rangle$, leading to different sequences of transitions: $|\Psi_i\rangle \to |\Psi_{\kappa_1}\rangle \to |\Psi_{\kappa_2}\rangle \to \cdots \to |\Psi_f\rangle$.

A subtle aspect of this treatment is that T_{fi} vanishes exactly at the resonant frequency $\Omega_{\mathfrak{L}} = \omega_a/m + (m-1)\omega_b/m$, due to quantum interference among different excitation pathways [12]. However, this conclusion is fundamentally altered when some higher-order quantum electrodynamics (QED) effects are taken into account. These effects introduce finite widths for the excited levels.

To incorporate these effects, one adds an imaginary component to each of the excited-level energies: $\varepsilon_e^a \to \varepsilon_e^a + i\Gamma/2$ and $\varepsilon_e^b \to \varepsilon_e^b + i\Gamma/2$, where Γ is the (natural) width of the excited levels, assumed identical for both atoms for simplicity. With this modification, T_{fi} no longer vanishes, and quantum interference is found to contribute a suppression factor to $|T_{fi}|^2$, expressed as $\frac{\Gamma^2/4\hbar^2}{(\Omega_{\mathfrak{L}}-\omega_b)^2}[f_o(m)]^2$, where $f_o(m)$ is a function of m with values approximately in the range (1/m, 1). It is also noteworthy that the exact cancellation of T_{fi} in standard perturbation theory no longer holds when $\Omega_{\mathfrak{L}}$ deviates from the exact resonant frequency. In such a detuned case, the MPMA process incorporates an inseparable subprocess of photon emission by the m-atom system. Again, quantum interference leads to a suppression factor analogous to that observed in the case of exact resonance [12].

Accounting for this quantum interference suppression factor, the transition rate can be approximately estimated as:

$$W_{mpma} = \frac{1}{2^{4m+1}\pi^{3m-1}\hbar} (\Omega_{\mathfrak{L}}/\omega_a) (\Omega_{\mathfrak{L}}/\omega_b)^{m-1} n_{\lambda^3}^m \frac{\Gamma^2 \tau_a \tau_b^{m-1} \Omega_{\mathfrak{L}}^m}{(\Omega_{\mathfrak{L}} - \omega_b)^{2m}} [f(m)]^{2m} \rho(E_f). \tag{5}$$

In this expression, τ_a represents a decay rate parameter of the A-species atom, defined as $\tau_a = 4\alpha_e \omega_a^3 |\langle e_a|\hat{\mathbf{d}}_a|g_a\rangle|^2/3e^2c^2$, where e is the charge of the electron, c is the speed of light and $\alpha_e = e^2/4\pi\hbar c\epsilon_0 \approx 1/137$ is the fine-structure constant. Similarly, $\tau_b = 4\alpha_e \omega_b^3 |\langle e_{b_0}|\hat{\mathbf{d}}_{b_0}|g_{b_0}\rangle|^2/3e^2c^2$ is a decay rate parameter of one B-species atom. The term n_{λ^3} represents the number of laser photons in a volume of $\lambda^3 = (2\pi c/\Omega_{\mathfrak{L}})^3$, i.e., $n_{\lambda^3} = N_{\gamma}\lambda^3/V$ with V being the volume of the laser field. Finally, f(m) is a function of m with values roughly in the range (1/m, 1) [14].

In [12], we argued for the existence of a characteristic length, l_{mpma} , which plays a fundamental role in the cooperative excitation of an m-atom system. This excitation can only

occur when the system's linear size is smaller than l_{mpma} . A key mechanism determining the value of l_{mpma} arises from the uncertainty principle in quantum mechanics. In this m-th order process, while energy conservation is maintained between the initial and final quantum states of the joint system, all intermediate virtual quantum transitions involve energy non-conservation. The largest energy mismatch among the m intermediate virtual quantum transitions, denoted Δ_{ε} , is given by $\hbar |\Omega_{\mathfrak{L}} - \Omega_{a}|$. The uncertainty principle implies that these intermediate states can last a duration $\Delta t = \hbar/2\Delta_{\varepsilon}$. Consequently, a corresponding length, $l_{\Delta} \equiv c\Delta t$, emerges. It is natural to propose that $l_{mpma} = \alpha l_{\Delta}$, where α is a constant less than unity. Some experimental evidence [3, 4] supporting this analysis of l_{mpma} is discussed in [12].

To optimize the MPMA process for certain applications, it could be crucial to achieve a large value of l_{mpma} and a small value of Δ_{ε} . This can be more effectively realized by utilizing two laser fields for the joint excitation of the m-atom system [12]. One laser, denoted \mathfrak{L}_1 , is intended to excite the A-species atoms and has a frequency $\Omega_{\mathfrak{L}_1}$ that differs from ω_a by a small amount $\delta_a = |\Omega_{\mathfrak{L}_1} - \omega_a|$. The second laser, denoted \mathfrak{L}_2 , has a frequency $\Omega_{\mathfrak{L}_2}$ close to ω_b . The frequencies are configured to satisfy the relation:

$$\Omega_{\mathfrak{L}_1} + (m-1)\Omega_{\mathfrak{L}_2} = \omega_a + (m-1)\omega_b. \tag{6}$$

With this setup, the m-atom system can undergo joint excitation by absorbing one photon from the \mathcal{L}_1 laser and m-1 photons from the \mathcal{L}_2 laser. The transition rate can be analyzed straightforwardly, taking a form similar to the right side of Eq. (5) and involving the intensities of both lasers. In this two-laser scenario, $\Delta_{\varepsilon} = \hbar \delta_a$, which can be widely tuned. The potential smallness of δ_a is generally constrained by factors such as the natural width of excited states and the thermal Doppler width of the atom gas. In some cases, δ_a can reach the order of $2\pi \times 10^{10}$ Hz, allowing l_{mpma} to be as large as 3.0 mm, which is around four orders of magnitude larger than the wavelength of visible light. In the remainder of this paper, we will not differentiate whether the MPMA process involves one or two laser fields, as the formalisms and estimations are quite similar.

According to Eq. (5), the transition rate for an m-atom system is inherently low, reflecting its nature as a high-order QED process. However, this rate applies to a single m-atom system. When considering the entire atomic gas, the overall transition rate for a specific atom can be significantly enhanced, driven by the key fact that a vast number of m-atom systems

involving this atom are subject to excitation transitions simultaneously and in parallel.

Consider an atomic gas with a linear size l_{gas} and a homogeneous density of B-species atoms denoted by ρ_b . The total number of A-species atoms is denoted by \mathcal{N}_A and the total number of B-species atoms is \mathcal{N}_B (assuming $\mathcal{N}_A < \mathcal{N}_B/m$). Select one A-species atom, labeled A_o , and denote the total number of B-species atoms that can be jointly excited with the A_o atom as N_{bo} . If $l_{gas} < l_{mpma}$, then $N_{bo} = \mathcal{N}_B$. In the case where $l_{gas} > l_{mpma}$, N_{bo} can be approximated by $\rho_b l_{mpma}^3$, with a factor assumed to be unity for simplicity. The total number of m-atom systems that can be formed by the A_o atom and N_{bo} B-species atoms is roughly $C_{m-1}^{N_{bo}} = \frac{N_{bo}(N_{bo}-1)...(N_{bo}-(m-1))}{(m-1)(m-2)...1} \approx N_{bo}^{m-1}/(m-1)!$. According to quantum mechanics, each of these m-atom systems is subject to an excitation transition, and the overall transition rate for the a_o atom is given by:

$$W_{a_o} \approx \frac{N_{bo}^{m-1}}{(m-1)!} W_{mpma}. \tag{7}$$

This significant quantum enhancement can be clearly illustrated through the analysis of relevant wavefunctions. For simplicity, consider a separate subsystem of the atomic gas composed of the A_o atom and N_{bo} B-species atoms with a linear size smaller than l_{mpma} . At t=0, with all atoms in their ground states, the wavefunction of the system (comprising the A_o atom, N_{bo} B-species atoms, and the laser field) is $\Psi_{ini} = |g_{a_o}\rangle|g_{b_1}\rangle|g_{b_2}\rangle\dots|g_{b_{N_{bo}}}\rangle|N_{\gamma}\rangle$. For a specific choice of m-1 atoms out of N_{bo} B-species atoms indexed by k_1, k_2, \dots, k_{m-1} $(k_1 < k_2 < \dots < k_{m-1})$, the quantum state with the chosen m-atoms being excited (by absorbing m laser photons) is:

$$\Psi_{k_1, k_2, \dots, k_{m-1}}^{ex} = |e_{a_o}\rangle |e_{b_{k_1}}\rangle |e_{b_{k_2}}\rangle \dots |e_{b_{k_{m-1}}}\rangle |\Psi_{k_1, k_2, \dots, k_{m-1}}^b\rangle |N_{\gamma} - m\rangle$$
(8)

Here, $|e_{b_{k_i}}\rangle$ (for $i=1,2,\ldots,m-1$) denotes the excited state of the k_i -th B-species atom, and $|\Psi^b_{\overline{k_1,k_2,\ldots,k_{m-1}}}\rangle$ represents the quantum state of the remaining B-species atoms in their ground states.

At a small time t, the quantum state $\Psi(t)$ is given by:

$$\Psi(t) = c_0(t)\Psi_{ini} + \sum_{k_1, k_2, \dots, k_{m-1}} c_{k_1, k_2, \dots, k_{m-1}}(t)\Psi_{k_1, k_2, \dots, k_{m-1}}^{ex}.$$
(9)

According to perturbation theory, $|c_{k_1,k_2,\dots,k_{m-1}}(t)|^2 \approx W_{mpma}t$. The probability of the a_o atom remaining unexcited is:

$$P_{unex}(t) = |c_0(t)|^2 = 1 - \sum_{k_1, k_2, \dots, k_{m-1}} |c_{k_1, k_2, \dots, k_{m-1}}(t)|^2 \approx 1 - \frac{N_{bo}^{m-1}}{(m-1)!} W_{mpma} t.$$
 (10)

The rate of change of $P_{unex}(t)$ with respect to t corresponds to the total transition rate. At a time τ_{ex} corresponding to the inverse of W_{a_o} , $P_{unex}(\tau_{ex})$ deviates significantly from unity, indicating a high probability that the A_o atom has undergone an MPMA transition.

Combining Equations (5), and (7), one finds,

$$W_{a_o} \approx \frac{1}{2^{4m+1}\pi^{3m-1}\hbar} n_{\lambda^3}^m \frac{\Gamma^2 \tau_a \tau_b^{m-1} \Omega_{\mathfrak{L}}^m}{(\Omega_{\mathfrak{L}} - \omega_b)^{2m}} [f(m)]^{2m} \frac{N_{bo}^{m-1}}{(m-1)!} \rho(E_f). \tag{11}$$

Consider a homogeneous gas system where $N_{bo} \approx \rho_b l_{mpma}^3$. According to Eq. (11), W_{a_o} depends critically on N_{bo} , and thus on l_{mpma} . In certain scenarios, W_{a_o} can become exceptionally large when l_{mpma} is significant. For instance, consider $l_{mpma} \approx l_{\Delta} \approx 1.0$ mm, corresponding to $\Omega_{\mathfrak{L}} - \omega_a$ on the order of $2\pi \times 10^2$ GHz, then $N_{bo} \approx 10^{12}$ with $\rho_b = 10^{15}$ atoms/cm³. Assuming τ_a and τ_b are on the order of $2\pi \times 10$ MHz, $\Gamma \approx \hbar \tau_b$, $\rho(E_f) \approx 10^{-3}/\hbar \tau_a$, and $n_{\lambda^3} \approx 10^{-2}$, W_{a_o} exceeds 10^{20} s⁻¹ for m=4, an exceptionally high value. This suggests that a regulatory mechanism may be necessary to prevent W_{a_o} from becoming excessively large.

In fact, there exists a fundamental mechanism which regulates the values of l_{mpma} and W_{a_o} . The timescale $T_{a_o} \equiv 1/W_{a_o}$, represents the average duration for the A_o atom to complete the excitation transition. As this transition is part of a collective process involving an m-atom system, the entire system must transition within the same timescale T_{a_o} . As a result, the m-atom system should be confined within a distance smaller than $l_{a_o} \equiv cT_{a_o} = c/W_{a_o}$, ensuring that collective excitation complies with the principle that information does not propagate faster than the speed of light. Notably, most m-atom systems involving the A_o atom have a size close to the maximum allowed length l_{mpma} , while those with significantly smaller sizes are negligible in number. This introduces a further constraint: $l_{mpma} \leq \beta l_{a_o}$, where β is a constant less than unity. This condition is generally satisfied when W_{a_o} is small but becomes more restrictive as W_{a_o} increases.

This analysis leads to two scenarios for determining l_{mpma} :

- i) If $\alpha l_{\Delta} < \beta l_{a_o}$: l_{mpma} can be directly set to αl_{Δ} , and W_{a_o} can be computed using Eq. (11).
- ii) If $\alpha l_{\Delta} > \beta l_{a_o}$: since $l_{mpma} \leq \beta l_{a_o}$ is required, l_{mpma} can no longer equal αl_{Δ} , Instead, it is adjusted to a reduced value (see Fig. 2), which decreases W_{a_o} and increases l_{a_o} , yielding the following self-consistent relationship:

$$l_{mpma} = \beta l_{ao} = \beta c / W_{ao}. \tag{12}$$

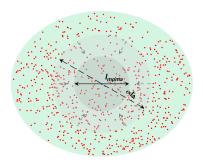


FIG. 2. In a dense gas, the characteristic length l_{mpma} of an MPMA process can self-adjust to values below αl_{Δ} , ensuring that relativistic causality is not violated.

Using Equations (11) and (12), along with $N_{bo} = \rho_b l_{mpma}^3$, it becomes possible to consistently determine l_{mpma} and W_{a_o} . This regulating mechanism significantly influences the value of l_{mpma} , ensuring a more physically reasonable value for W_{a_o} and preventing it from becoming unnaturally large.

This self-regulating mechanism is a fundamental and intriguing aspect of the MPMA process. As a result, W_{a_o} cannot be increased arbitrarily but likely tends to exhibit a near-saturation behavior below a scale of 10^9 s⁻¹ or so. Unlike other saturation behaviors in the field of matter-light interaction, the near-saturation of W_{a_o} is governed by the principle of relativistic causality. This distinctive feature highlights the profound interplay between quantum mechanics and the principles of relativity, offering a glimpse into their interconnected nature.

A fundamental application of the MPMA process is to enhance ultraweak atomic absorption transitions to observable levels. For instance, consider an electric multipole absorption, where the absorbed electric 2^j -pole (Ej) photon [15] carries an angular momentum of $j\hbar$ (j=2,3,...). General analysis indicates that the transition rate scales approximately as $(a/\lambda)^{2j}$ [15], where a is the linear size of the atom and λ is the wavelength of the photon involved [15]. Typically, $(a/\lambda) \approx 10^{-4}$, so the E2 transition has a strength on the order of 10^{-8} compared to the electric dipole (E1) transition. Similarly, the E3 transition is on the order of 10^{-16} , the E4 transition on the order of 10^{-24} , and so forth. For an E4 transition, the transition rate is typically around 10^{-16} s⁻¹ [16], making it extremely challenging to observe. Higher multipole transitions, such as E5 and E6, are generally undetectable in their unenhanced form. However, the MPMA process can be used to elevate these ultraweak transition rates to levels comparable to E1 transitions, enabling experimental investigation

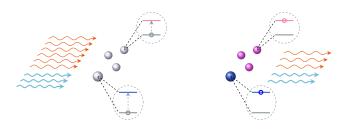


FIG. 3. Schematic illustration of an Ej-photon-absorption process involving a simultaneous joint excitation of a four-atom system. The A'-species atom (represented by the large ball) absorbs an Ej photon (thick blue lines), while each of three B-species atoms absorbs a laser photon (red lines). of these otherwise elusive transitions.

The integration of the MPMA process into an atomic Ej-photon absorption transition of a certain species of atoms, denoted as A'-species atoms, can generally be accomplished by adjusting the frequencies of the stimulating (laser) photons to satisfy energy conservation in a joint transition process involving a corresponding m-atom system. In this process, one A'-species atom absorbs an Ej-photon, while simultaneously, m-1 B-species atoms are excited by absorbing m-1 laser photons (see Fig. 3 for an example with m=4). The laser frequency $\Omega_{\mathfrak{L}_2}$ is close to, but not equal to, ω_b , and the energy conservation is expressed as:

$$\hbar\Omega_{Ej} + (m-1)\hbar\Omega_{\mathfrak{L}_2} = \hbar\omega_{a,Ej} + (m-1)\hbar\omega_b, \tag{13}$$

where Ω_{Ej} is the frequency of the Ej photon, and $\omega_{a,Ej}$ denotes the Ej-transition frequency of an A'-species atom. Although the transition rate for this MPMA process within a single m-atom system is extraordinarily small—partly due to the extremely small strength of the Ej transition in the A'-species atom—the overall transition rate for a given A'-species atom in a two-species atomic gas can become significant. This overall rate benefits from an enhancement factor of N_{bo}^{m-1} , which can surpass many orders of magnitude, rendering the rate substantial enough to reach an observable level.

A flux of Ej photons is essential for experimentally investigating these enhanced atomic absorption transitions. Technologically, twisted light [17–21], such as Laguerre–Gaussian and Bessel beams, offers a feasible and natural solution, as it delivers photons carrying high angular momentum. The interaction between twisted photons and atoms has been theoretically studied [22–24], but experimental observation remains an open challenge. One major difficulty arises from the inherently low transition rates of multipole transitions, as discussed above. Recently, a quadrupole transition in a trapped 40 Ca⁺ ion has been observed

via the absorption of a twisted photon [25]. With MPMA enhancement, atomic absorption of twisted photons could become significantly more detectable. Furthermore, it would be interesting to explore whether MPMA enhancement can also be utilized to facilitate the emission of high-multipole photons from atomic gases.

In summary, MPMA processes represent a fundamental and fascinating aspect of matterlight interactions, exhibiting unusual behaviors such as the near-saturation of the atomic transition rates under certain circumstances. MPMA processes can be used to amplify ultraweak atomic transitions by many orders of magnitude, offering a powerful tool for probing atomic phenomena that would otherwise remain inaccessible to experimental observation. The super-enhancement mechanism underlying MPMA processes is deeply rooted in the fundamental principles of quantum mechanics, holding substantial potential to enrich our understanding of light-matter interactions and advance our comprehension of quantum collective phenomena.

REFERENCES

- C. L. Cowan, F. Reines, F. B. Harrison, H. W. Kruse, and A. D. McGuire, Science 124, 103 (1956).
- [2] W. Kaiser and C. G. B. Garrett, Phys. Rev. Lett. 7, 229 (1961).
- [3] J. C. White, Optics Letters **6**, 242 (1981).
- [4] E. Pedrozo-Peñafiel, R. R. Paiva, F. J. Vivanco, V. S. Bagnato, and K. M. Farias, Phys. Rev. Lett. 108, 253004 (2012).
- [5] J. R. Leite and C. B. D. Araujo, Chemical Physics Letters 73, 71 (1980).
- [6] D. L. Andrews and M. Harlow, The Journal of Chemical Physics 78, 1088 (1983).
- [7] M. H. Nayfeh and G. B. Hillard, Physical Review A 29, 1907 (1984).
- [8] M. S. Kim and G. S. Agarwal, Physical Review A 57, 3059 (1998).
- [9] A. Muthukrishnan, G. S. Agarwal, and M. O. Scully, Physical Review Letters **93**, 093002 (2004).
- [10] Z. Zheng, P. L. Saldanha, J. R. R. Leite, and C. Fabre, Physical Review A 88, 033822 (2013).

- [11] C. Hettich, C. Schmitt, J. Zitzmann, S. Kühn, I. Gerhardt, and V. Sandoghdar, Science 298, 385 (2002).
- [12] Y. Yu, ChinaXiv: 202503.00168.
- [13] Formally, $\rho(E_f) = \delta(E_f \varepsilon_e^a (m-1)\varepsilon_e^b)$. However, due to the intrinsic coupling between the excited atoms and the quantum electromagnetic field, the excited levels of each atom exhibit a resonant-state nature and possess a finite width (see, e.g., [26]). As a result, the m-atom system also acquires a finite width at $E_f = \varepsilon_e^a (m-1)\varepsilon_e^b$, and the density of states could be approximated as: $\rho(E_f) \sim \frac{\Gamma_{eff}/2}{(E_f \varepsilon_e^a (m-1)\varepsilon_e^b)^2 + (\Gamma_{eff}/2)^2}$, where Γ_{eff} is comparable to the maximum natural widths of the quantum states $|e_a\rangle$ and $|g_{b_i}\rangle$, differing by a factor between unity and m. Moreover, thermal broadening of the excited states can be incorporated into $\rho(E_f)$ as well.
- [14] The term $f_o^2(m)$ in the suppression factor associated with quantum interference has been incorporated into $[f(m)]^{2m}$ in Eq. 5.
- [15] E. M. L. V. B. Berestetskii and L. P. Pitaevskii, Quantum Electrodynamics, 2nd ed. (Elsevier (Singapore) Pte Ltd., Singapore, 2008).
- [16] The rate here can refer to either the spontaneous E4-photon emission rate or the E4-photon absorption rate. When it refers to the E4-photon absorption rate of 10^{-16} s⁻¹, it should be noted that the requisite E4-photon field possesses a certain strength, corresponding to a photon density where the number of photons within a volume of λ^3 —where λ is the wavelength of the E4 photon—is approximately on the order of 10^{-7} .
- [17] L. Allen, M. J. Padgett, and M. Babiker, in *Progress in Optics*, Vol. 39, edited by E. Wolf (Elsevier, 1999) pp. 291–372.
- [18] A. M. Yao and M. J. Padgett, Advances in Optics and Photonics 3, 161 (2011).
- [19] A. Zannotti, F. Diebel, M. Boguslawski, and C. Denz, Advanced Optical Materials 5, 1600629 (2016).
- [20] P. S. J. Russell, R. Beravat, and G. K. L. Wong, Philosophical Transactions of the Royal Society A 375, 20150440 (2017).
- [21] M. Babiker, D. L. Andrews, and V. E. Lembessis, Journal of Optics 21, 013001 (2019).
- [22] A. Afanasev, C. E. Carlson, and A. Mukherjee, Phys. Rev. A 88, 033841 (2013).
- [23] H. M. Scholz-Marggraf, S. Fritzsche, V. G. Serbo, A. Afanasev, and A. Surzhykov, Phys. Rev. A 90, 013425 (2014).

- [24] S. A.-L. Schulz, A. A. Peshkov, R. A. Müller, R. Lange, N. Huntemann, C. Tamm, E. Peik, and A. Surzhykov, Phys. Rev. A 102, 012812 (2020).
- [25] C. Schmiegelow, J. Schulz, H. Kaufmann, T. Ruster, U. Poschinger, and F. Schmidt-Kaler, Nature Communications 7, 12998 (2016).
- [26] J. D.-R. C. Cohen-Tannoudji and G. Grynberg, Atom-Photon Interactions: Basic Processes and Applications (John Wiley & Sons, 1998).